



## Nitrous oxide emissions from a beech forest floor measured by eddy covariance and soil enclosure techniques

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Abstract

Spring time soil nitrous oxide (N<sub>2</sub>O) fluxes were measured in an old beech (*Fagus syl-*  
*vatica* L.) forest with eddy covariance (EC) and chamber techniques. The aim was to  
compare the two techniques and to test whether EC can be used in the trunk-space of  
the forest to measure N<sub>2</sub>O. Mean N<sub>2</sub>O fluxes over the five week measurement period  
were 5, 10 and 16 μg N m<sup>-2</sup> h<sup>-1</sup> from EC, automatic chamber and manual chambers,  
respectively. When data from one hot spot chamber was excluded the mean N<sub>2</sub>O flux  
of 8 μg N m<sup>-2</sup> h<sup>-1</sup> from the soil chambers nearly equaled to the mean flux of 7 μg  
N m<sup>-2</sup> h<sup>-1</sup> measured with EC from the direction where soil chambers were located. Spatial  
variability in the N<sub>2</sub>O emissions was high in soil chamber measurements, while the EC  
integrated over this spatial variability and suggested that N<sub>2</sub>O emissions were uniform  
within the footprint area. The highest emissions measured with the EC occurred dur-  
ing the first week of May when the trees were leafing and when soil moisture content  
was at its highest. To our knowledge, this is the first study to demonstrate that the EC  
technique can be used to measure N<sub>2</sub>O fluxes in the trunk-space of a forest. If cham-  
ber techniques are used to estimate ecosystem level N<sub>2</sub>O emissions from forest soils,  
placing of the chambers should be considered carefully to cover the heterogeneity in  
the soil N<sub>2</sub>O emissions.

1. Introduction

Microbial activity in soil ecosystems is the major source of nitrous oxide (N<sub>2</sub>O) to the  
atmosphere. Nitrous oxide acts as a greenhouse gas in the troposphere accounting  
for approximately 6% of the radiative forcing of all greenhouse gases. In addition,  
N<sub>2</sub>O takes part in ozone depleting reactions in the stratosphere. Long atmospheric life  
time, 120 years, and a global warming potential of about 300 times higher than carbon  
dioxide, in 100-years horizon, makes N<sub>2</sub>O an important factor in the global climate  
system (IPCC 2001).

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Forest soils are a source of  $\text{N}_2\text{O}$  to the atmosphere but the source strength of different forests is still uncertain. Annual emissions range from near 0 to 20 kg of  $\text{N}_2\text{O}$ -N per hectare, depending on atmospheric N deposition, forest type and management practices (Schmidt et al., 1988; Tietema et al., 1991; Papen and Butterbach-Bahl, 1999; Bowden et al., 2000; Beier et al., 2001). Several soil physical, chemical and biological factors control microbial  $\text{N}_2\text{O}$  production in the soil. In forest ecosystems, soil moisture, soil temperature and nitrogen availability are the key factors regulating  $\text{N}_2\text{O}$  emissions (Butterbach-Bahl et al., 2002; Schindlbacher et al., 2004; Papen and Butterbach-Bahl, 1999). Increase in soil moisture, temperature or the availability of mineral nitrogen usually stimulates soil microbial processes and consequent  $\text{N}_2\text{O}$  production.

High spatial and temporal variability characterizes  $\text{N}_2\text{O}$  emissions from different ecosystems (Ambus and Christensen, 1995; Christensen et al., 1996; Röver et al., 1999; Yanai et al., 2003). In agricultural ecosystems this spatial and temporal variability results from small scale differences or changes in soil nitrate and ammonium, and organic material contents (Röver et al., 1999; Yanai et al., 2003). This heterogeneity and changes in soil variables controlling  $\text{N}_2\text{O}$  emissions make it challenging to reliably estimate  $\text{N}_2\text{O}$  emissions in ecosystem level.

The most commonly used technique in  $\text{N}_2\text{O}$  emission measurements is the closed chamber technique (see e.g. Papen and Butterbach-Bahl, 1999; Schulte-Bisping and Brumme, 2003). It has the advantages of being relatively easy and inexpensive to use, and it is especially appropriate when soil fluxes are related to the chemical and microbiological factors of the soil in small scale. However, soil chambers are prone to problems such as: possible modification of the flow at the soil-air interface; representativeness of the sampling places; and the disturbance of the chamber collars to the soil ecosystem (see e.g. Hutchinson and Livingston, 2001; Pumpanen et al., 2003; Savage and Davidson, 2003). As opposed to the chamber techniques, the micrometeorological techniques do not disturb the soil, and the fluxes are usually measured continuously allowing information on the temporal variation of fluxes to be obtained (Fowler and Duyzer, 1989). The most direct micrometeorological flux measurement method, the

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EC method, relies on the measurement of variations in vertical wind velocity and trace gas concentration above the source surface with high time resolution (see e.g. Baldocchi, 2003). The EC technique integrates fluxes over a large source area, providing a tool for ecosystem level flux measurements.

The EC method is routinely used to measure fluxes of for example carbon dioxide (CO<sub>2</sub>) and water vapor (H<sub>2</sub>O) above vegetation canopies. It has recently also been adopted for trunk-space measurements. Baldocchi et al. (1986) conducted the first EC studies of soil-atmosphere CO<sub>2</sub> exchange below forest canopies. According to them the requirements for sub-canopy EC measurements are: steady state conditions, no sources and sinks between the soil surface and measurement height, and an extended level and horizontally homogeneous upwind fetch (see Baldocchi and Meyers 1991).

Simultaneous EC and chamber measurements of N<sub>2</sub>O fluxes have been conducted on agricultural grassland ecosystems, but data from forest ecosystems is lacking (Smith et al., 1994; Cristensen et al., 1996). The spatial variability of N<sub>2</sub>O emissions from forest ecosystems has only been addressed using chamber techniques (Ambus and Christensen, 1995; Butterbach-Bahl et al., 2002). To our knowledge, this is the first study to compare EC and chamber techniques to measure N<sub>2</sub>O fluxes from a forest floor. The aims of this study were 1) to evaluate whether the EC technique can be used below a forest canopy to measure soil emissions of N<sub>2</sub>O, 2) to compare the magnitude and variability of N<sub>2</sub>O fluxes measured by EC and chamber techniques, and 3) to obtain data on the N<sub>2</sub>O emissions from forest soils.

The measurements took place during a five week field measurement campaign FOXNOTE (Forest Oxidized Nitrogen Transport Experiment). The experiment was part of the EU project NOFRETETE and took place in an old beech (*Fagus sylvatica* L.) forest in Sorø, Denmark. During the five week campaign N<sub>2</sub>O emissions were measured with manual and automatic chambers, and using the EC technique in the trunk-space of the beech forest. To link the N<sub>2</sub>O emissions to environmental parameters, soil extractable mineral nitrogen content, soil temperature and soil moisture, and meteorological parameters were also measured.

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2. Materials and methods

2.1. Site description

The experiment was conducted in Denmark in the forest Lille Bøgeskov (Small Beech-forest) near Sorø on the island of Zealand (55°29' N, 11°39' E). The forest is located in a flat terrain and covers about 1.5 square kilometers of mainly of 82 year old beech (*Fagus sylvatica* L.) trees. Approximately 200 m on the south-east of the measurement site there is a small plantation of Norway spruce (*Picea abies* L.). The campaign period extended from 2 May to 5 June 2003. The average tree height of beech trees is 25 m and diameter is 38 cm, and the stand density is about 283 stems ha<sup>-1</sup> (Pilegaard et al., 2003). Total Leaf Area Index above the measurement height was 5.2 m<sup>2</sup> m<sup>-2</sup> on 26–27 May 2003, as measured with an LAI 2000 Plant Canopy Analyzer (LI-COR, Lincoln, Nebraska, USA). The soil in the area is either Alfisol or Mollisol according to the American Soil Taxonomy system, and it has a pH of 4 to 5 and a 10–40 cm deep organic layer with a C/N ratio of about 20 in the upper organic layers and about 10 in the lower mineral layers. A detailed description of the site is given in Pilegaard et al. (2003), and the placing of soil chambers, EC measurement system and soil sampling places are shown in Fig. 1.

2.2. Eddy covariance measurement system

The eddy covariance (EC) measurement system consisted of an ultrasonic 3-D anemometer (Solent 1012, Gill Ltd., Lymington, Hampshire, UK) and a tunable diode laser (TDL) trace gas analyzer (TGA100, Campbell Scientific Inc., Logan, Utah, USA). The TDL system consists of a temperature and current controlled single mode diode laser, tuned to an IR N<sub>2</sub>O absorption band, mounted in a liquid nitrogen dewar. Concentration measurement is achieved by passing the infra red laser beam through absorption cells containing sample and reference gases. The reference gas (2000 ppm N<sub>2</sub>O) is drawn through the short reference cell under same temperature and pressure

conditions as the sample air. As the length of the reference cell is only 4 cm, as compared to the length of the sample tube of 1.5 m, a high concentration of the reference gas is needed to obtain a clear absorption peak for the peak tracking algorithm. The concentration in the sample air ( $C_s$ ) is determined from the ratio of the reference and sample absorbances as follows:

$$C_s = \frac{C_r L_r R}{L_s(1 - R) + L_{ss}}, \quad (1)$$

where  $C_r$  is the concentration of the reference gas, and  $L_s$ ,  $L_r$  and  $L_{ss}$  are the lengths of the absorption tube, and short reference and sample cells, respectively.  $R$  is the ratio of the correlation of the sample and reference absorbance functions to the covariance of the reference gas filtered absorption functions (Edwards et al., 2003).

In the measurement setup the fluxes of  $N_2O$  were measured below the forest canopy at 3.0 m height. A Busch rotary-vane pump (RB0021-L) was used to draw the sample air to the TDL analyzer. The sample air was first passed through a diffusive drier (PD1000, Perma pure Inc.) to remove excess water vapor that could infer the analysis. Total flow rate of the air entering the drier was  $17 \text{ l/min}^{-1}$ , from which the sample flow was 14 and the purge flow was  $3 \text{ l/min}^{-1}$ , adjusted with a needle valve and a flow meter attached to the bottom of the dryer, respectively. Sample air leaving the dryer was directed to the TDL analyzer via a 10 m long Teflon tubing with i.d. of 4 mm. During the measurement period, pressure inside the sample cells was kept at approximately 70 mbar and the measurements were done at 10 Hz frequency.

### 2.3. Eddy covariance data processing

The vertical flux of the  $N_2O$  is calculated as the covariance between the vertical wind velocity ( $w$ ) and the  $N_2O$  concentration. Averaging time for flux calculations was 30 min and linear de-trending of concentrations was done prior to flux calculations.

Since trunk-space EC measurements lack a standard criterion for removing low turbulence periods, such as  $u^*$  criterion used for above canopy measurements, we filtered

the flux data using a criterion for standard deviation of vertical wind speed. We removed the measurements with standard deviation of vertical wind speed less than  $0.07 \text{ m s}^{-1}$ , which was seen as suitable for trunk-space EC measurements at a pine forest site in Finland (data not shown). Also erroneous data caused by electronics were removed.

5 This data filtering removed approximately 78% of the night time (22:00–08:00) data points and about 35% of the day time (08:00–22:00) data points. During the first two weeks of the campaign, the loss of data was smaller than during the rest of the campaign when on occasional days almost all of the night time data was removed due to low turbulence.

10 The Lagrangian stochastic trajectory calculation procedure (Thomson, 1987) was used for estimation of flux footprint functions. The simulations were performed releasing  $3 \times 10^4$  particles from the ground and followed until the upwind distance from the observation point accounted over 99% of the total flux. A one and half order closure model for neutral stratification by Massman and Weil (1999) was used to parameterize  
15 flow statistics within the canopy. A detailed description of the model used is given by Markkanen et al. (2003) and Rannik et al. (2000).

#### 2.4. Flux detection limit of the eddy covariance system

The detection limit of the EC measurement system depends on the signal noise of the TDL instrument ( $\sigma_c$ ) and the standard deviation of vertical wind speed ( $\sigma_w$ ) at the  
20 observation level. Flux detection limit ( $\sigma_x$ ) was calculated as

$$\sigma_x = \frac{\sigma_w \sigma_c}{\sqrt{Tf}} \quad (2)$$

where  $T$  is the averaging time and  $f$  the measurement frequency. The noise level of TDL for  $\text{N}_2\text{O}$  ( $\sigma_c$ ) has been estimated to be 1 ppb, and the typical standard deviation of vertical wind speed ( $\sigma_w$ ) below forest canopy at 3 m height is about  $0.15 \text{ m s}^{-1}$ . For a  
25 30 min averaging period with 10 Hz measurement frequency the detection limit of the

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$\text{N}_2\text{O}$  flux ( $\sigma_x$ ) at Sorø measurement site is  $4.6 \mu\text{g N m}^{-2} \text{ h}^{-1}$ . For daily mean values of  $\text{N}_2\text{O}$  emissions, the detection limit decreases to approximately  $1 \mu\text{g N m}^{-2} \text{ h}^{-1}$ .

## 2.5. Soil enclosure measurements

Enclosure measurements were conducted with six manual and one automatic static chamber located north to north-west from the EC mast (Fig. 1). The manual static chamber collars, made of 30 cm diameter and 15 cm long PVC pipes, were pushed ca. 5 cm depth into the soil giving a headspace volume of  $7.1 \text{ dm}^3$ . At the time of gas sampling the chamber collars were closed with Perspex lids equipped with butyl rubber stoppers. Four gas samples were taken at 20 min intervals from the headspace by syringe and needle through the stopper. The manual chamber measurements were conducted on weekly basis. Automatic chamber measurements were conducted with an automated gas sampling system (UIT, Dresden, Germany). A 10 cm high stainless steel chamber frame covering an area of  $0.7 \times 0.7 \text{ m}^2$  was pushed 5 cm into the soil. During measurements the automatic chamber was sealed by a 10 cm high chamber box sliding on horizontal bars and three gas samples were taken at 40 min intervals. The automatic chamber was operated in 3-hour intervals during 7 to 14 May, and thereafter twice a day. All chambers had been in place at least 16 weeks prior to the campaign. The gas samples from both manual and automatic chamber systems were injected to 3.5 ml pre-evacuated glass vials (Venojects®) until analysis by a Shimadzu gas chromatograph 14B (Shimadzu, Kyoto, Japan) equipped with Electron Capture Detector, and an automatic headspace sampler (Mikrolab, Århus, Denmark).

## 2.6. Soil measurements

Soil samples were collected on daily basis from 6 to 14 May and thereafter twice a week. Soil samples were collected from the top 10 cm layer with a 2.5 cm diameter soil core; six samples from the area close to manual soil chambers, and six samples from an area adjacent to the EC measurement system (Fig. 1). In total 12 soil samples

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per each sampling day were obtained and frozen on the day of sampling. The soils were melted at +4°C and sampled for gravimetric soil moisture (105°C; 24 h) and soil extraction with 1M KCl (1:5 w vol<sup>-1</sup>). Soil extracts were immediately frozen and later analyzed for nitrate (NO<sub>3</sub>) and ammonium (NH<sub>4</sub>) with aBran+Luebbe AutoAnalyzer 3 System (Bran+Luebbe, Norderstedt, Germany). In the field, soil temperature at 2 and 10 cm depths (Pt-100, Risø National Laboratory, Denmark) and soil volumetric moisture content at 10 cm (TDR, ThetaProbe ML2x, Delta-T) were measured continuously.

## 2.7. Statistical analysis

Eddy covariance and chamber techniques were compared during one week of intensive measurements with the automated chamber (7 to 14 May), and separately on 7 and 15 May when both automatic and manual chambers were operated. Daily means of the fluxes between the automatic chamber and the EC technique were compared using T-test for independent samples (SPSS 12.01, SPSS Inc.). Daily fluxes from all the soil chambers and from the EC, on 7 and 15 of May, were tested using the non-parametric Kruskal-Wallis test. To test wind direction dependency of the EC fluxes, the flux data was divided into eight 45° wind direction sectors (see Fig. 1), and the fluxes from the wind sectors were compared using a non-parametric Kruskal-Wallis test.

## 3. Results

### 3.1. The magnitude and variability of N<sub>2</sub>O emissions

The co-spectra of the N<sub>2</sub>O concentration and vertical wind velocity from the EC/TDL system can be used to assess the frequency behavior of the measured N<sub>2</sub>O flux (Kaimal and Finnigan, 1994). An example of N<sub>2</sub>O-*w* co-spectra, calculated using two hours of data from the afternoon of 25 May is shown in Fig. 2. The frequencies around 2×10<sup>-3</sup>–2×10<sup>-2</sup> Hz make the highest contribution to the fluxes measured at this time.

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The wind speed measured below the canopy was  $0.27 \text{ m s}^{-1}$ . The shape of the spectrum does not follow the theoretical above canopy co-spectrum, but is in line with the below canopy co-spectrum of  $\text{CO}_2$  and  $w$ , and  $\text{H}_2\text{O}$  and  $w$  measured in the trunk-space of a pine forest in Finland (data not shown). As the frequency dependence of the fluxes measured below canopies is less pronounced than the classical  $f^{-4/3}$  behavior, this leads to a relatively higher contribution of high frequency, or small scale, eddies to the measured flux.

Average  $\text{N}_2\text{O}$  emissions measured with EC, automatic chamber and manual chambers during the measurement period 2 May–6 June were  $5.4$ ,  $10.0$  and  $16.0 \mu\text{g N m}^{-2} \text{ h}^{-1}$ , respectively (Table 1). The 10% and 90% percentiles of the half hourly EC data ranged from  $-6.9$  to  $19.4 \mu\text{g N m}^{-2} \text{ h}^{-1}$  and in the automatic chamber data from  $6.0$  to  $15.0 \mu\text{g N m}^{-2} \text{ h}^{-1}$ . Daily variation in the  $\text{N}_2\text{O}$  fluxes, expressed as coefficients of variation, was approximately 10-fold higher in the EC data than in the automatic chamber data, and two-fold higher in the EC data than in the manual chamber data (Table 1).

Daily emissions measured with EC were on average  $9 \mu\text{g N m}^{-2} \text{ h}^{-1}$  during the first week of May and thereafter on average  $4 \mu\text{g N m}^{-2} \text{ h}^{-1}$  (Fig. 3a). The highest daily  $\text{N}_2\text{O}$  emissions of  $20 \mu\text{g N m}^{-2} \text{ h}^{-1}$  were measured on the first measurement day, 2 May (Fig. 3a). Low fluxes were measured between 14 and 24 of May after which the emissions peaked on again on 25 and 31 of May at  $8 \mu\text{g N m}^{-2} \text{ h}^{-1}$ .

Temporal variation in  $\text{N}_2\text{O}$  emission, measured by the automatic chamber, was smaller during the first half of the measurement period than in the last half of the period (Fig. 3b).  $\text{N}_2\text{O}$  emissions peaked on 12, 18, 25, 28 and 29 May with a maximum daily emission of  $17.7 \mu\text{g N m}^{-2} \text{ h}^{-1}$  on 28 May (Fig. 3b). Emissions with manual soil chambers exhibited high small scale spatial variability, illustrated by large SE error bars (Fig. 3b). One out of six manual chambers gave constantly higher emission values than the other five. If the data from this “hot spot” soil chamber was excluded the average  $\text{N}_2\text{O}$  emission over the whole measurement period was  $7.7 \mu\text{g N m}^{-2} \text{ h}^{-1}$ .

Comparison of the EC and automatic chamber fluxes during 7 to 14 of May shows that the EC exhibited higher temporal variation in  $\text{N}_2\text{O}$  fluxes than the automatic cham-

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ber (Fig. 3c). The automatic chamber measured on average higher fluxes than the EC. On three out of seven days, 7, 10 and 11 May, the fluxes from the automatic chamber were significantly higher than those from the EC measurements ( $p < 0.05$ , T-test). When comparing the fluxes from all the chambers (manual and automatic) to the fluxes from the EC, on 7 and 15 May, the chamber and EC fluxes did not differ significantly from each other ( $p = 0.27$ ).

### 3.2. Influence of soil environmental parameters on $N_2O$ emissions

Soil nitrate ( $NO_3$ ) content fluctuated very little during the measurement period (Fig. 4a). The two areas, close to the soil chambers and close to the EC mast, differed from each other with respect to soil  $NO_3$  content ( $p < 0.01$ ). In the area close to the chambers,  $NO_3$  content was on average 2.7 times higher than in the area close to the EC mast, and 1.5 times higher than the average of the whole area (Figs. 4a and b). Soil  $NO_3$  peaked on 12 May, after a rainfall event. The minimum  $NO_3$  content was measured on the same day as that of  $NH_4$  (19 May). Soil  $NH_4$  content during the measurement period was on average 9.3 mg N per kg of dry soil (Figs. 4a and 4b). Temporal variation in soil  $NH_4$  was larger than that of soil  $NO_3$ . Soil  $NH_4$  content peaked on the 8 May, decreased to a minimum on 19 May, and increased again at the end of May (Fig. 4b).

At the start of the measurement period, soil moisture and  $NH_4$  contents were at their maximum and soil temperature at its minimum (Figs. 4b and 4c). The decrease in soil moisture content throughout the measurement period was disrupted by several rainfall events. Soil surface temperature at 10 cm depth increased during the measurement period from approximately 7 °C to 12 °C (Fig. 4c). Fluctuation at the surface was greater than deeper in the soil (results not shown).

The  $N_2O$  emissions measured with the EC technique followed the pattern of soil  $NO_3$  and  $NH_4$  contents (Figs. 3a and 4a, 4b). A weak dependency of  $N_2O$  emissions on the soil  $NO_3$  and  $NH_4$  contents was found during a period 8 to 19 May (Fig. 5).  $N_2O$  emissions decreased as the soil N content decreased and increased again after the soil N contents increased in the end of May. The minimum  $N_2O$  emission was measured on

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the same day, 19 May, as the minimum in soil  $\text{NO}_3$  and  $\text{NH}_4$  contents. The highest  $\text{N}_2\text{O}$  emissions were measured on 2 May when soil moisture was at its highest (above 50% wfps) (Figs. 3a and 4c). The emissions peaked on 13 and on 25 May after a rainfall and consequent increase in soil moisture. Soil  $\text{N}_2\text{O}$  emissions correlated positively with soil moisture content from 8 to 19 May (Fig. 6).

### 3.3. Wind direction dependency of $\text{N}_2\text{O}$ fluxes

Nitrous oxide emissions measured by the EC were independent of wind direction. The average  $\text{N}_2\text{O}$  flux was above  $6 \mu\text{g N m}^{-2} \text{ h}^{-1}$  from all other wind directions, and  $2\text{--}4 \mu\text{g N m}^{-2} \text{ h}^{-1}$  from south east direction (Fig. 7), however, this difference was statistically insignificant ( $p=0.49$ ). Soil chambers were located north to north-west from the EC mast, a wind sector from which EC measured a mean  $\text{N}_2\text{O}$  emission of  $7 \mu\text{g N m}^{-2} \text{ h}^{-1}$  (Figs. 1 and 7). This flux value is less than the emission of  $16.0 \pm 5.7 \mu\text{g N m}^{-2} \text{ h}^{-1}$  measured by manual soil chambers (mean  $\pm$  SE). Excluding the “hot spot” manual chamber, discussed in Sect. 4.1, and adding the data from the automatic chamber, during the days when both automatic and manual chambers were measured, the mean  $\text{N}_2\text{O}$  flux from the soil chambers decreased to  $8.0 \pm 1.9 \mu\text{g N m}^{-2} \text{ h}^{-1}$ .

### 3.4. Diurnal variation in $\text{N}_2\text{O}$ emissions

There was no clear diurnal trend in  $\text{N}_2\text{O}$  emissions measured with EC or chamber techniques. Night time (22:00–08:00)  $\text{N}_2\text{O}$  emissions measured with the automatic chamber averaged to  $9.5 \mu\text{g N m}^{-2} \text{ h}^{-1}$ , and day time (08:00–22:00) emissions to  $10.3 \mu\text{g N m}^{-2} \text{ h}^{-1}$ . In the EC data, variation in  $\text{N}_2\text{O}$  fluxes was greater during day-time than during night-time. Due to low turbulence, 78% of the night time and 35% of the day time flux values were filtered out from the data, and the remaining data showed no daily pattern in the emission rate. Mean daily coefficient of variation of 26% in the automatic chamber data during 7 to 14 May, indicates very little temporal variability in  $\text{N}_2\text{O}$  emission at that specific chamber location (Table 1).

## 4. Discussion

We have shown that the EC measurement system can be placed in the trunk-space to measure  $N_2O$  emissions close to its sources, and that the EC and chamber techniques give comparable estimates of the  $N_2O$  fluxes. The facts supporting these findings are that 1) the emissions measured with the EC were comparable in magnitude and within the variability of the emissions measured by chambers, 2) the  $N_2O$  fluxes measured with EC responded to changes in environmental conditions, such as soil moisture and soil mineral N, in the same way as the emissions measured by chambers, and 3) the difference in the  $N_2O$  average emissions measured by the EC and chamber techniques can reasonably be attributed to the differences in the nitrogen availability in the soil at the EC flux footprint and in the area of chamber measurements.

Although, the EC  $N_2O$  fluxes were within the limits of mean  $\pm$  standard deviation given by the soil chambers, the  $N_2O$  emissions measured with the EC were on average lower than those measured with the chambers. When comparing the EC data to the data from the automatic chamber, the fluxes measured by the chamber were significantly higher than the fluxes measured by the EC. When the EC results were compared to all the soil chambers, the difference between the techniques was insignificant. Hence, the tests between the two techniques revealed that compared to the temporal variation the spatial variability in soil  $N_2O$  emissions is so high that it overrides the statistical significances of the mean tests. Also, it showed that the spatial variability in  $N_2O$  emissions is greater than the temporal variability in  $N_2O$  emissions for the study forest during this period. Christensen et al. (1996) compared EC and chamber techniques to measure  $N_2O$  emissions from an agricultural field. They measured 31 to 55% higher  $N_2O$  emissions with the EC technique than with the soil chambers. Still, this difference between the two techniques was within the uncertainty given by the spatial variability of the flux over the measurement area.

We measured much higher diurnal variation in  $N_2O$  fluxes with the EC than with the chambers (Table 1). As in EC the flux is a time average over the footprint area, part

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of this high variability can be explained by high spatial variability in the N<sub>2</sub>O fluxes. However, most of this variation in the EC data may also have resulted from random uncertainty when measuring fluxes close to the detection limit of the TDL instrumentation. According to Baldocchi and Meyers (1991), the EC technique is accurate but lacks precision when used near the forest floor. They concluded that the scatter in their studies was a combination of geophysical variability, and sampling and measurement errors, and that reliable data can be obtained by using long averaging times.

The highest N<sub>2</sub>O emissions with EC were measured during the first week of May. This week was rainy and the soil moisture content was at its highest, above 50% of water filled pore space (wfps). In such conditions, anaerobic microsites may have been created in the soil increasing N<sub>2</sub>O production from denitrification. Also, during this period when trees were leafing (the bud break period) the requirements for sub-canopy micrometeorological measurements were best fulfilled: the canopy of the beech forest remained open and allowed for greater turbulence in the trunk-space. The beech trees were fully leafed in the middle of May after which also turbulence intensity decreased. A short increase in mean wind speed inside the trunk-space was measured during the last measurement week, in June (data not shown).

In previous studies, based on manual chamber measurements, the average N<sub>2</sub>O emissions from annual emission measurements from this beech forest floor was 5 to 6  $\mu\text{g N m}^{-2} \text{ h}^{-1}$  (Ambus et al., 2001; Beier et al., 2001). Our spring time measurements of 5 to 16  $\mu\text{g N m}^{-2} \text{ h}^{-1}$  with EC and chambers are in line with this, and also with other emission measurements from temperate forest soils (Schmidt et al., 1988; Ambus and Christensen, 1995; Bowden et al., 2000).

According to Beier et al. (2001) this beech forest site has a closed nitrogen cycling, and only less than 5% of the nitrogen input to the ecosystem escapes the forest as gaseous nitrogen losses or leaching. Also, they found that soil NO<sub>3</sub> and NH<sub>4</sub> contents were similar in areas close to and between tree stems, indicating that the input of nitrogen to the forest floor via stemflow was insignificant to the soil pool of nitrogen. In our study, the soil NO<sub>3</sub> content varied between areas of the forest being higher in

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the area adjacent to the soil chambers than in the area close to the EC mast (Fig. 4a). As discussed above, manual soil chamber measurements showed a high small scale spatial variability in N<sub>2</sub>O emissions. This spatial variability can result from small scale differences in the availability of substrates for N<sub>2</sub>O production. Consequently, as the area close to the soil chambers had higher soil NO<sub>3</sub> content than the area adjacent to the EC mast, the lower N<sub>2</sub>O fluxes with EC may have resulted from constantly lower levels of soil substrates, such as NO<sub>3</sub>, for microbial N<sub>2</sub>O production. According to the footprint analysis, the area contributing 85% to the EC flux lies within 60 m from the EC mast, and the area contributing 50% to the eddy flux lies within 15 m around the measurement mast (Fig. 1). As the soil chambers were located between 40 to 50 m from the EC mast, they belong to the footprint area, but they contribute less to the eddy flux than the closest tens of meters from the eddy mast.

If in the future, ecosystem level emission estimates are based on chamber measurements only, the placement of soil chambers is critical in covering the spatial variability of the soil N<sub>2</sub>O emissions. Since the spatial variability was found to be greater than the temporal variability in the fluxes, the emphasis should be put on spatial coverage rather than high temporal resolution in the measurements. Aside with chamber measurements or as substitutive technique, ecosystem level flux measurements can be conducted with integrative techniques such as EC.

## 5. Conclusions

We have demonstrated that the EC technique can be used in the trunk space to measure N<sub>2</sub>O emissions from a forest floor. The fluxes measured by the EC and enclosure methods were comparable, although they measure at different spatial and temporal scales. Measurements with the chamber technique show a large spatial small scale variation in N<sub>2</sub>O fluxes. The EC technique integrates over a larger area covering both high and low microbial activity areas in the soil. The data suggests that if N<sub>2</sub>O emission measurements are based on chamber measurements only, the emphasis should

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be put on placement of soil chambers to cover the spatial variability in the soil N<sub>2</sub>O emission. Large scale integrative techniques, such as EC, can hence be the substitutive technique to estimate ecosystem level emissions of N<sub>2</sub>O.

*Acknowledgements.* European Commission through the project NOFRETETE (EVK2-CT2001-00106), Magnus Ehrnrooth foundation, Nordic Centre of Excellence, Research Unit BACCI and the Finnish Academy financially supported this work. We would also like to thank T. Pohja, E. Siivola and P. T. Sørensen for technical assistance.

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**Table 1.** Mean, median, minimum and maximum N<sub>2</sub>O emissions, and the coefficient of variation in the daily averaged eddy covariance (EC), automatic chamber and manual chamber measurements.

Method	N <sub>2</sub> O flux $\mu\text{g N m}^{-2} \text{ h}^{-1}$				Daily coeff. of var. mean (CV%) <sup>a</sup>
	Mean	Median	Min	Max	
EC daily mean	5.3	4.2	−0.8	20.3	264
Automatic chamber	10.0	9.2	0.2	29.4	26
Manual chamber	16.0	8.4	−4.3	93.5	141

<sup>a</sup>(stdev/mean)100%

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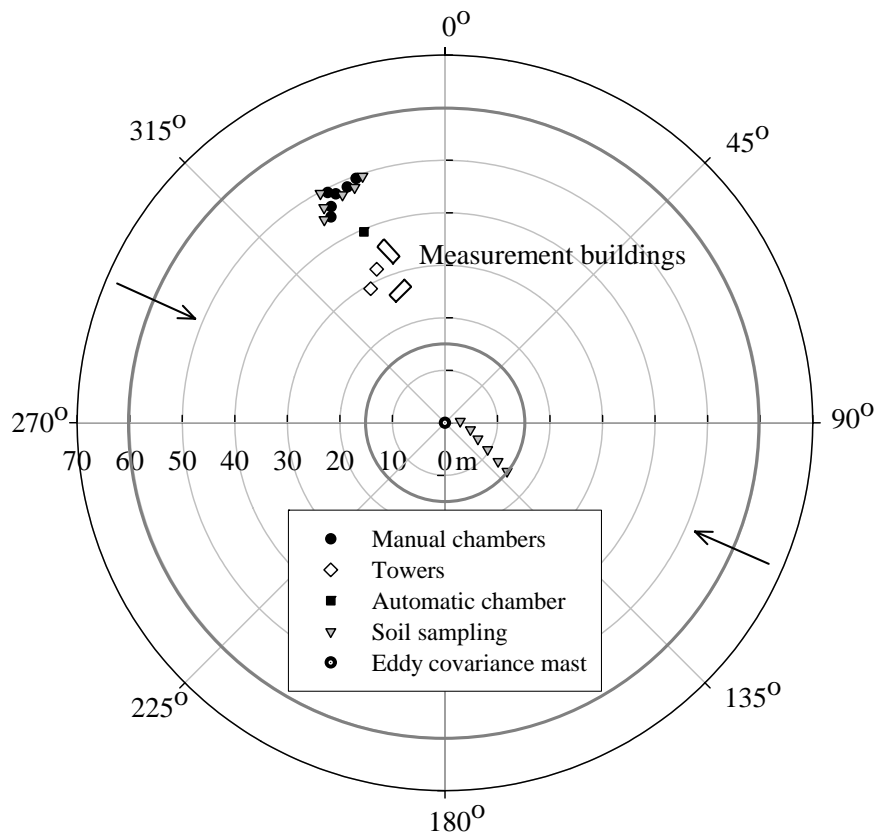
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**Fig. 1.** Site map of the beech forest Lille Bøgeskov. Black open circle in the origin represents the eddy covariance mast, black circles the manual chambers, black square the automatic chamber, grey triangles the soil sampling places, and open squares the two measurement towers and measurement buildings at the site. Grey lines around the site represent the footprint areas from which 50% (at 15 m) and 85% (at 60 m) of the  $\text{N}_2\text{O}$  fluxes originate. Arrows indicate prevailing wind directions during the campaign.

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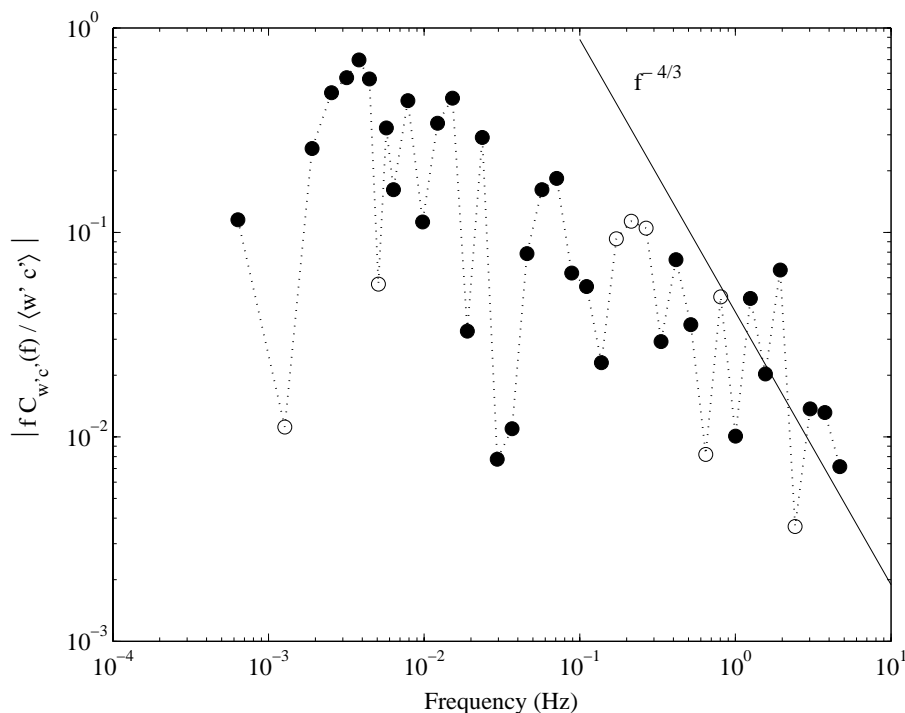
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**Fig. 2.** Co-spectra of the  $\text{N}_2\text{O}$  concentration and vertical wind velocity. The solid dots are frequencies with positive flux and open dots the frequencies with negative flux. The solid line indicates theoretical co-spectral behavior above canopy, where the co-spectra depends on the frequency to the power of  $-4/3$ .

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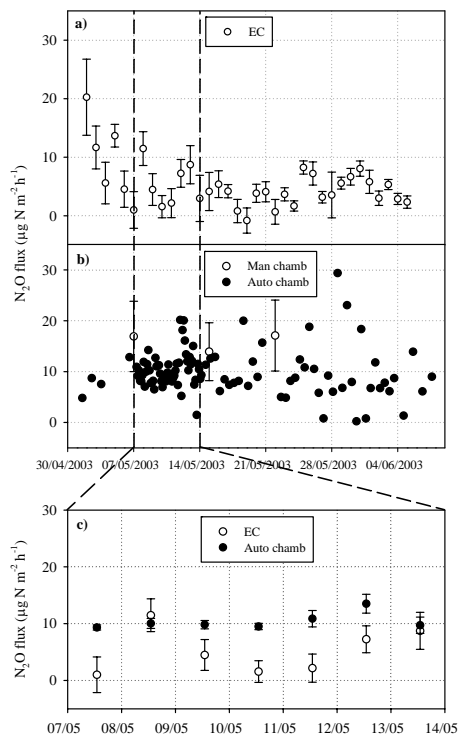
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**Fig. 3.** Nitrous oxide emissions from an old beech forest measured by eddy covariance (EC) and chamber techniques. **(a)** Open circles indicate EC daily mean N<sub>2</sub>O emissions  $\pm$  standard error of the mean ( $n_{EC}=3-44$ ), **(b)** Closed circles indicate N<sub>2</sub>O emission measured by an automatic chamber (Auto chamb), and open circles indicate N<sub>2</sub>O emissions measured by manual soil chambers (Man chamb). Error bars denote for  $\pm$  standard error of the mean ( $n_{chambers}=6$ ), **(c)** Comparison of daily means of the N<sub>2</sub>O flux measured by EC (open circles) and automatic chamber (closed circles) during 7 to 14 May. Error bars expresses  $\pm$  standard error of the mean ( $n_{EC}=12-38$ ,  $n_{chambers}=5-8$ ).

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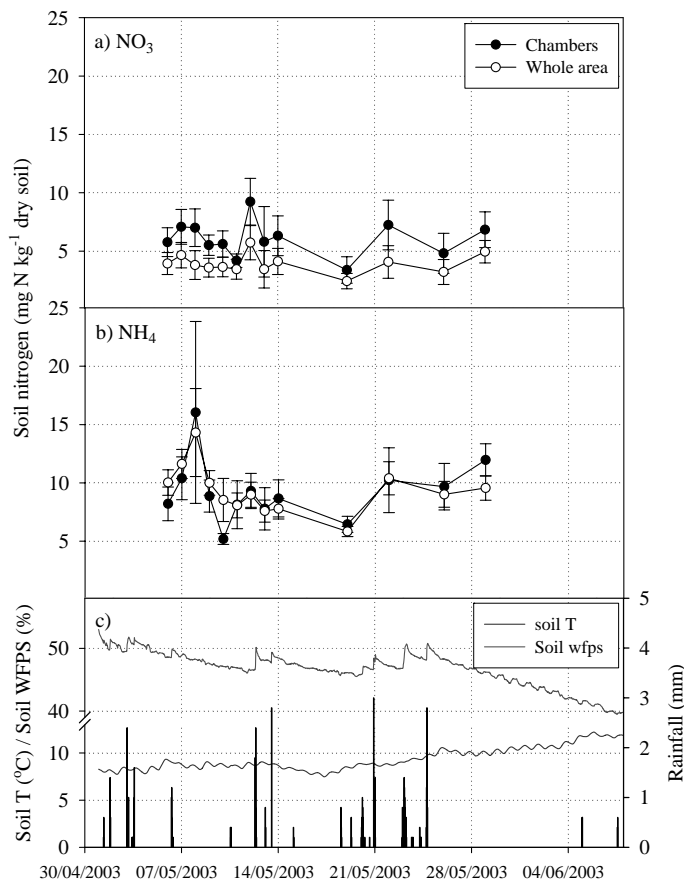
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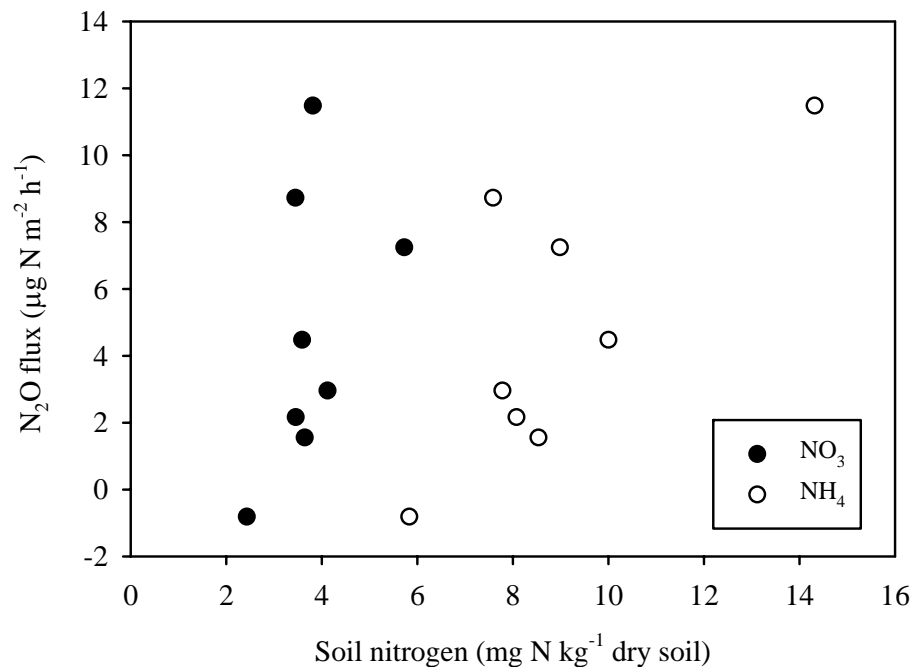


**Fig. 4.** Soil mineral nitrogen content at 0–10 cm depth of the soil from two areas: close to the soil chambers (black circles), and as an average over all the soil sampling places (open circles). **(a)**  $\text{NO}_3^-$ , **(b)**  $\text{NH}_4^+$ , **(c)** soil temperature at 10 cm, soil moisture as percentage of water filled pore space (WFPS%) at 0–6 cm, and rainfall.

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**Fig. 5.** Dependency of daily  $\text{N}_2\text{O}$  emissions measured by the EC on soil mineral nitrogen ( $\text{NO}_3^-$  and  $\text{NH}_4^+$ ) content during 8 to 19 May 2003.

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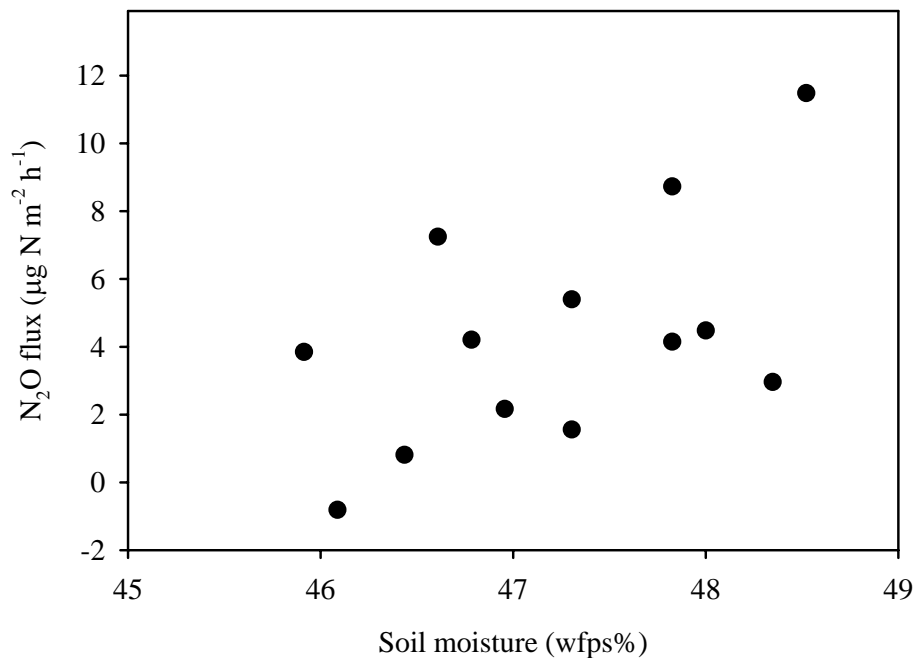
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**Fig. 6.** Dependency of daily N<sub>2</sub>O emissions measured by the EC on soil moisture (wfps%) during 8 to 19 May 2003.

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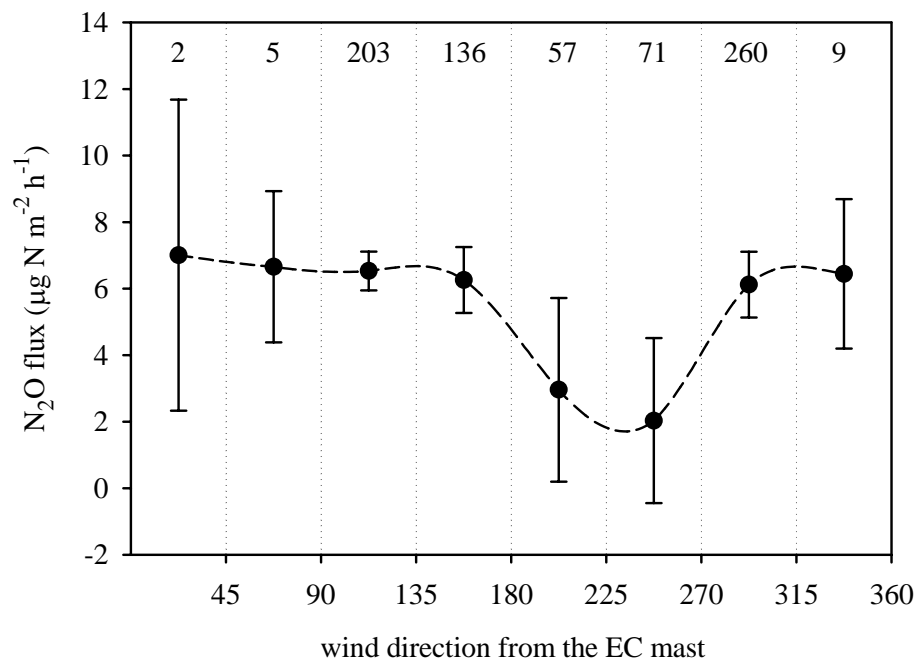
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**Fig. 7.** Nitrous oxide emissions from different wind direction sectors from the eddy covariance (EC) measurement mast. Black dots represent the mean N<sub>2</sub>O emission from each of 45° sector, error bars stand for  $\pm$  standard error of the mean ( $n=2\text{--}260$ ) and the dashed line is drawn to guide the eye of the reader. Numbers in the top of the figure stand for the number of data points from each wind sector.

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